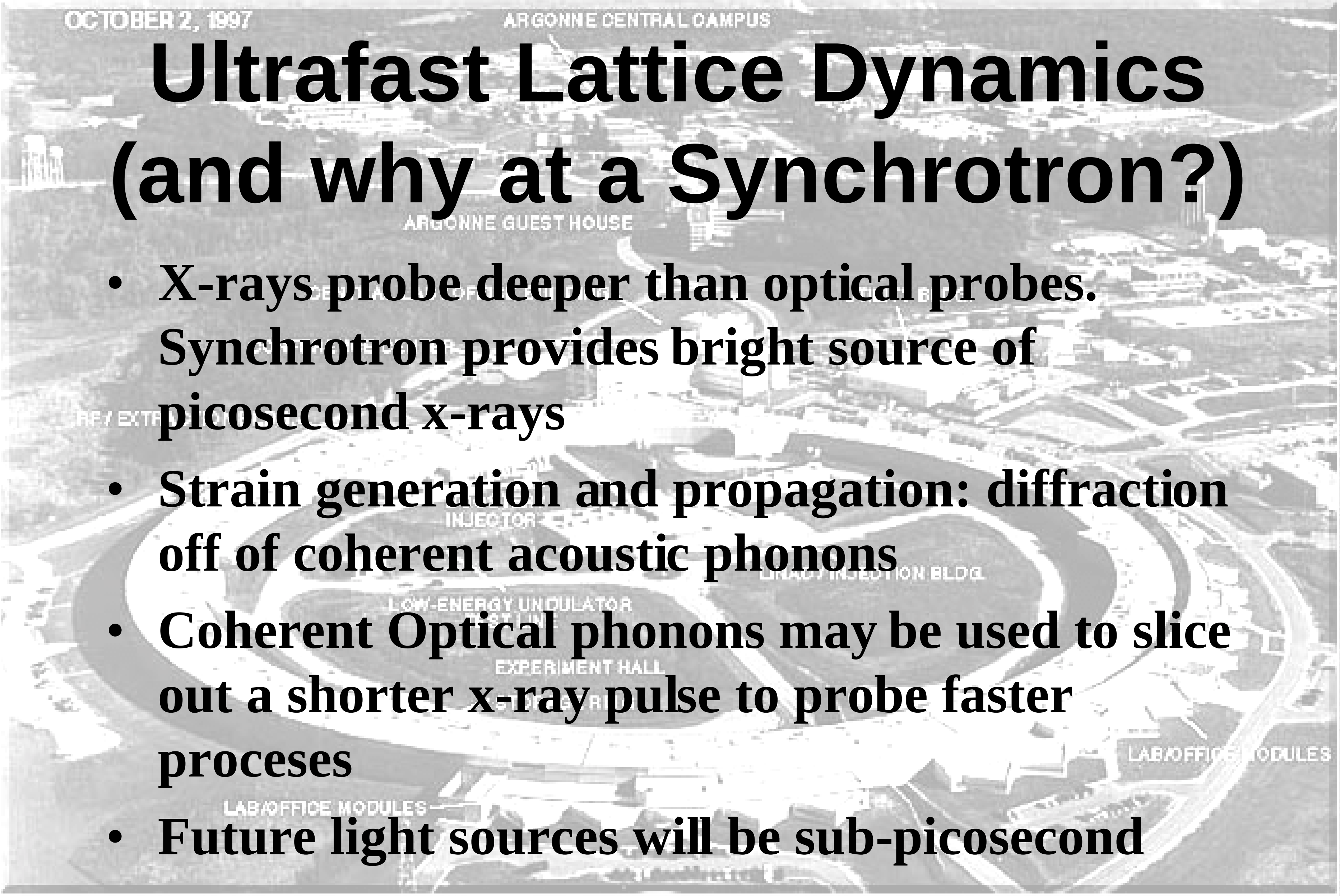
Ultrafast Lattice Dynamics (and why at a Synchrotron?)

- X-rays probe deeper than optical probes. Synchrotron provides bright source of picosecond x-rays
- Strain generation and propagation: diffraction off of coherent acoustic phonons
- Coherent Optical phonons may be used to slice out a shorter x-ray pulse to probe faster proceses
- Future light sources will be sub-picosecond



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ADVANCED PHOTON SOURCE
BEAM ACCELERATION & STORAGE SYSTEM

A. ELECTRON GUN

B. ELECTRON LINEAR ACCELERATOR
200 MeV-650 MeV

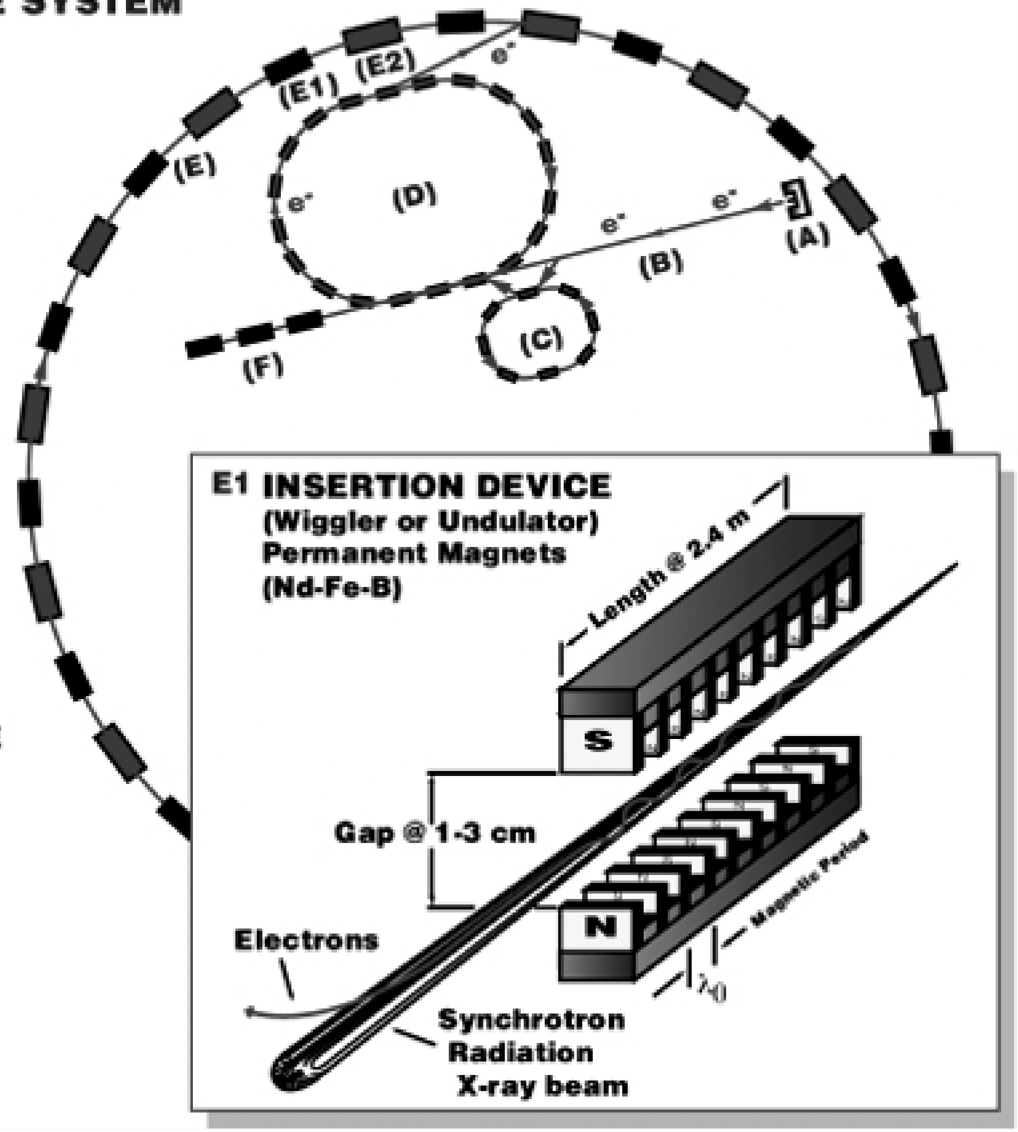
C. ACCUMULATOR RING

D. BOOSTER SYNCHROTRON
7 GeV

E. STORAGE RING
7 GeV nominal energy
E1 INSERTION DEVICE
E2 BENDING MAGNET

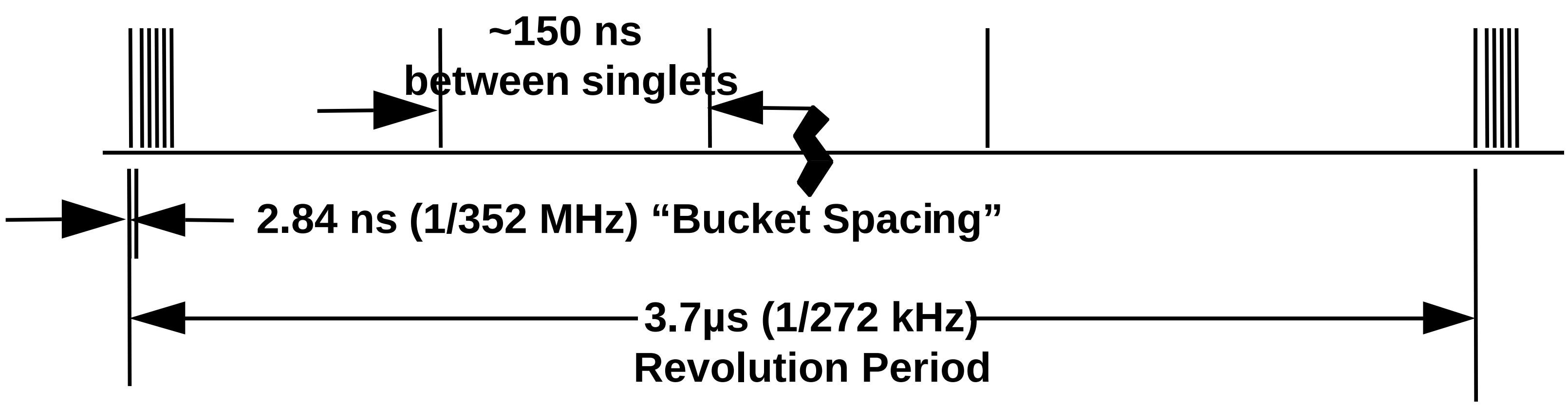
F. LOW-ENERGY UNDULATOR TEST LINE
400-650 MeV

NOTE: Diagrams not to scale

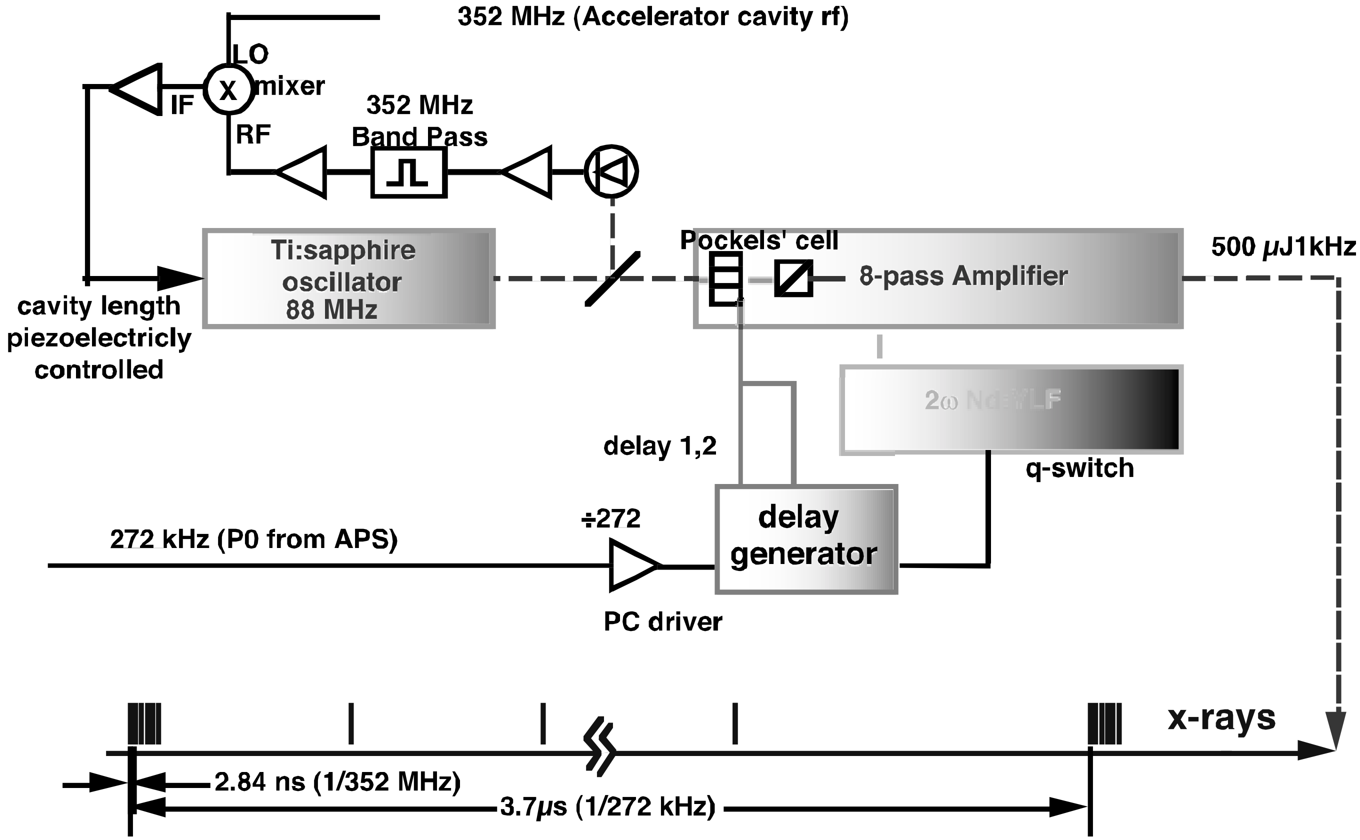


Electron/X-Ray Beam

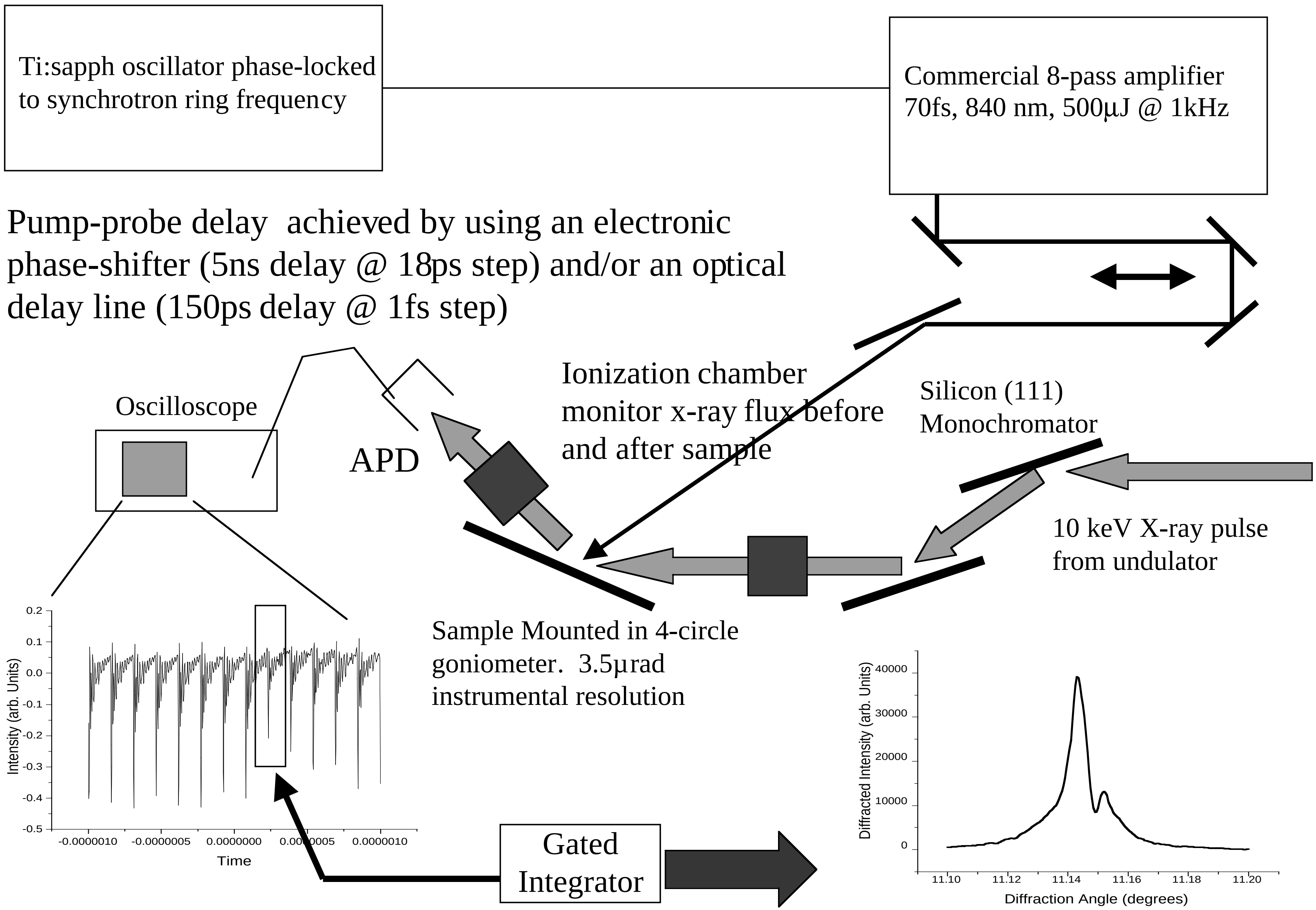
6 + 22 "singlets" bunch pattern with 100 mA ave current
Each bunch 50–100ps in width, 10 nC of charge, 7GeV
10^6 10 kV, x-rays/ bunch after monochromator (~10^{12} /s)

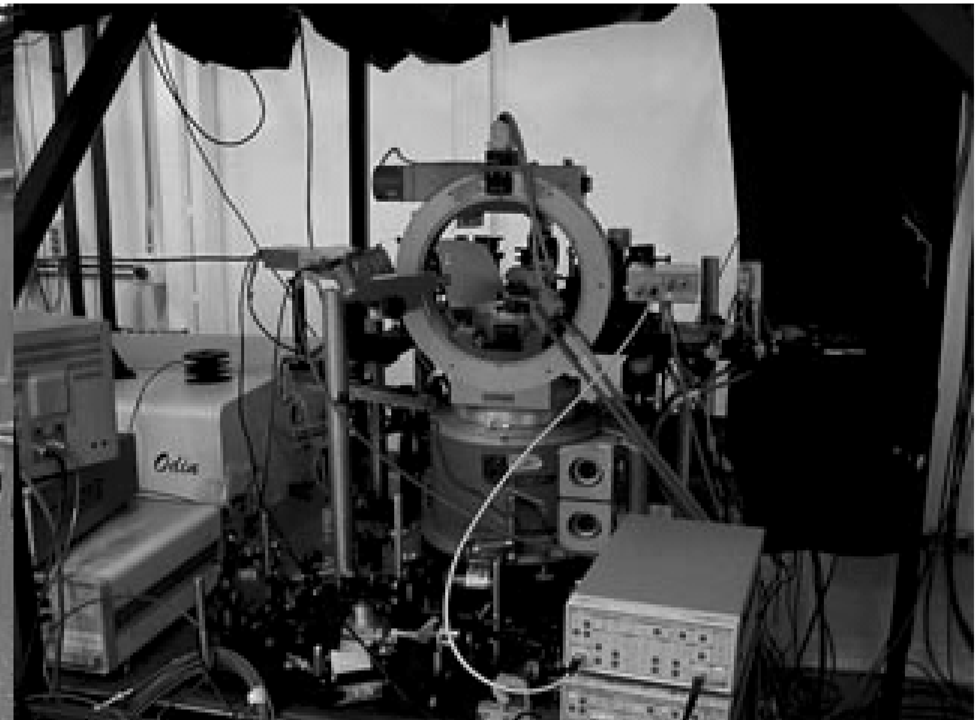


Laser–X-ray Timing



Pump-probe time-resolved x-ray diffraction



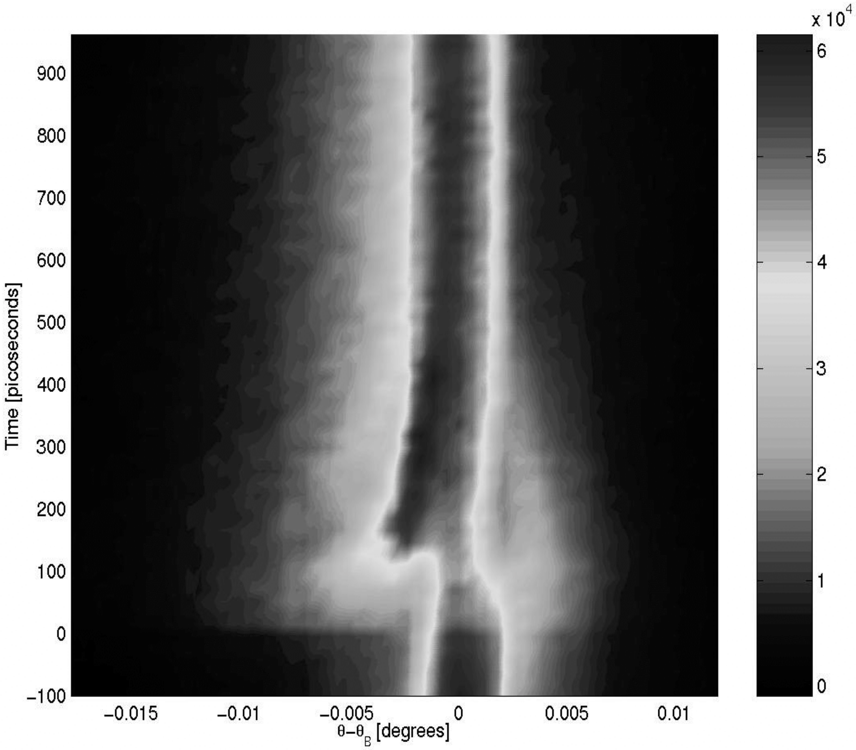

Odin


7 ID-D

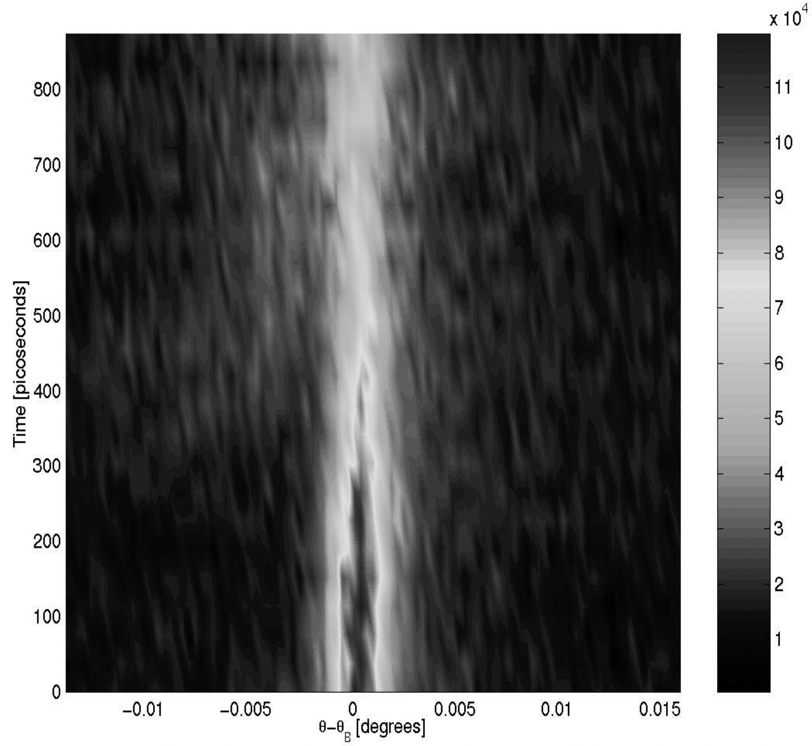
Experimental Data for Single Crystal InSb

Incident Laser: 10-12 mJ/cm^2, 70fs, 830 nm

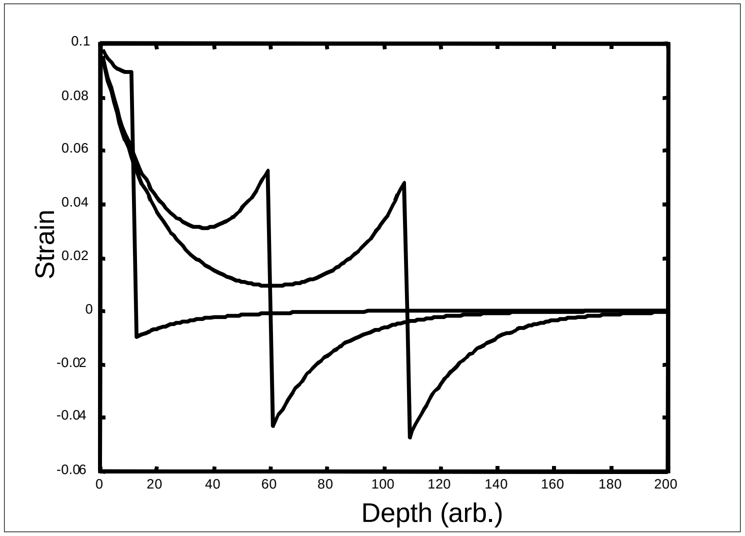
InSb (111)



InSb (222)



Generation and Detection of Coherent Acoustic Phonons

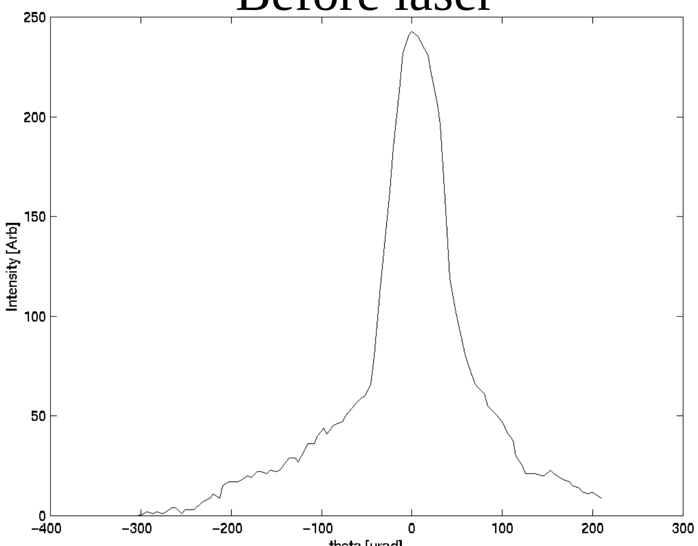


An ultrafast laser heats the material differentially across the penetration depth.

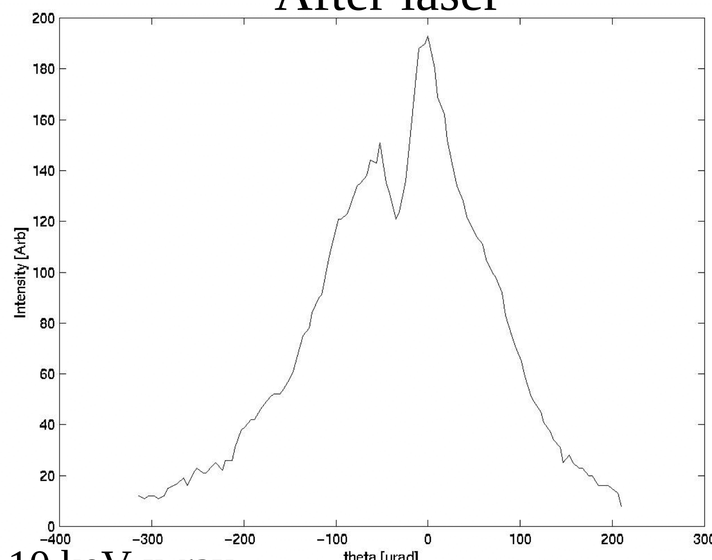
This causes a strain to propagate into the material at the speed of sound. Because the laser is impulsive the phonons are coherent over a wide spectrum.

Bragg diffraction is a sensitive probe to the strain.

Before laser

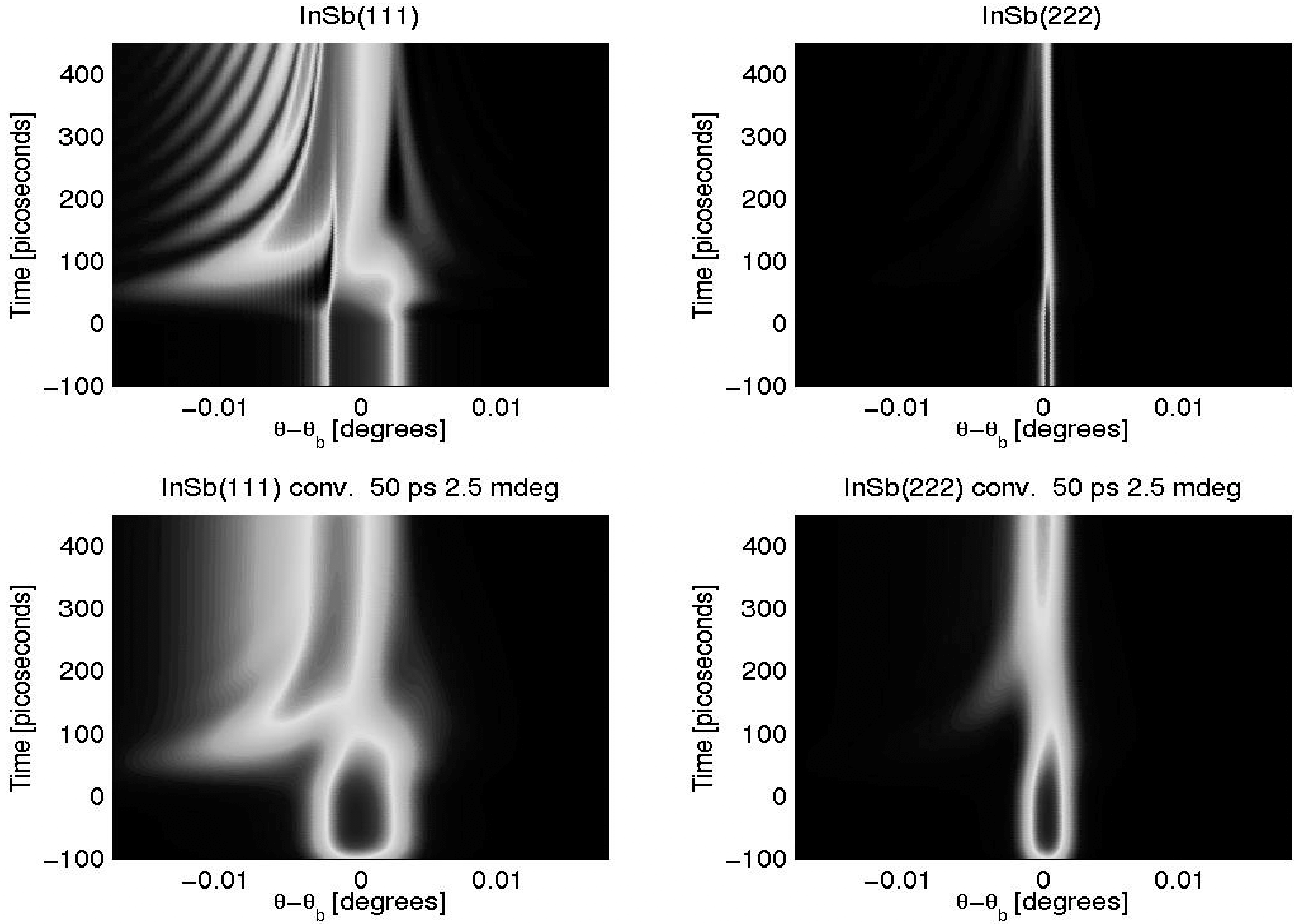


After laser



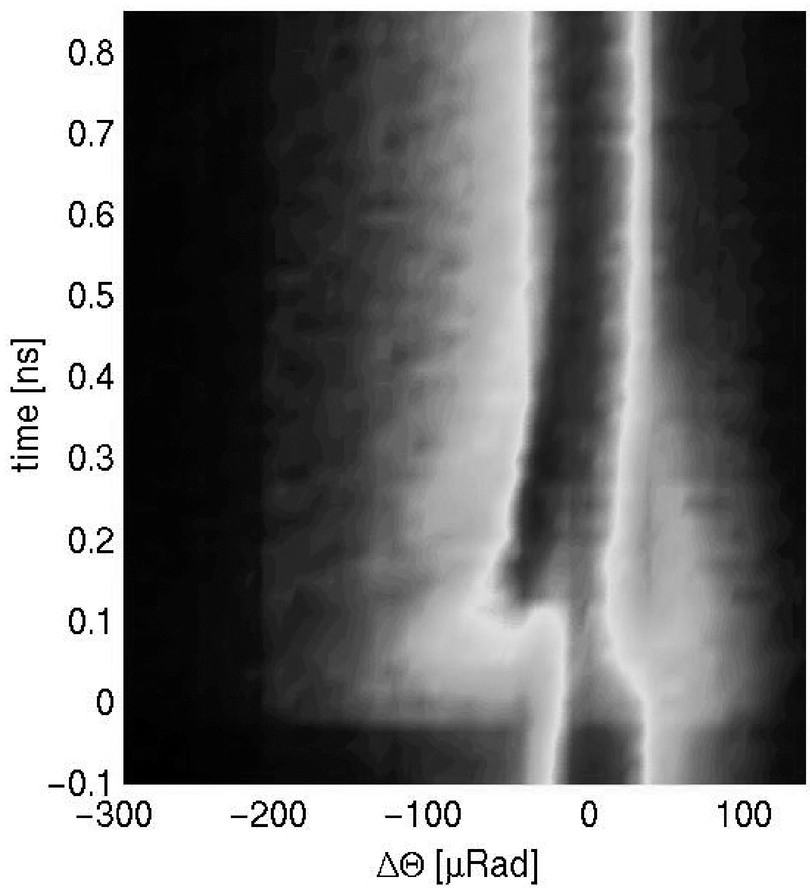
InSb (111) rocking curves, 10 keV x-ray

Dynamical Diffraction Simulation using Thomsen Strain Model

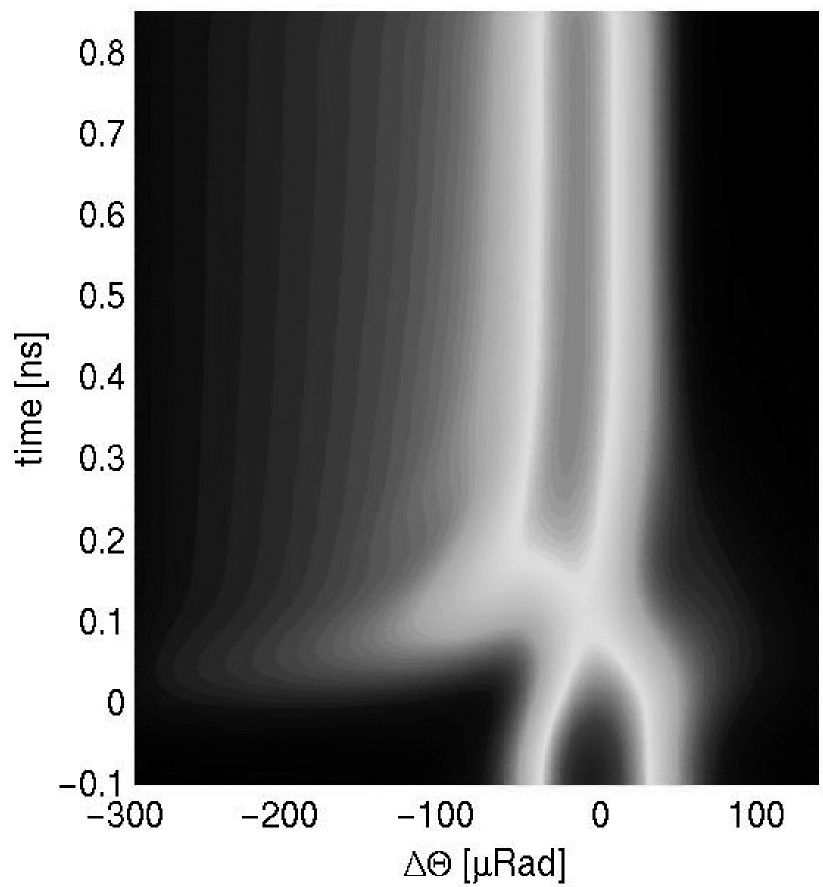


Probing coherent acoustic phonons in (111) InSb near the melt threshold

Experiment:
10 mJ/cm^2 incident fluence
10 keV X-rays



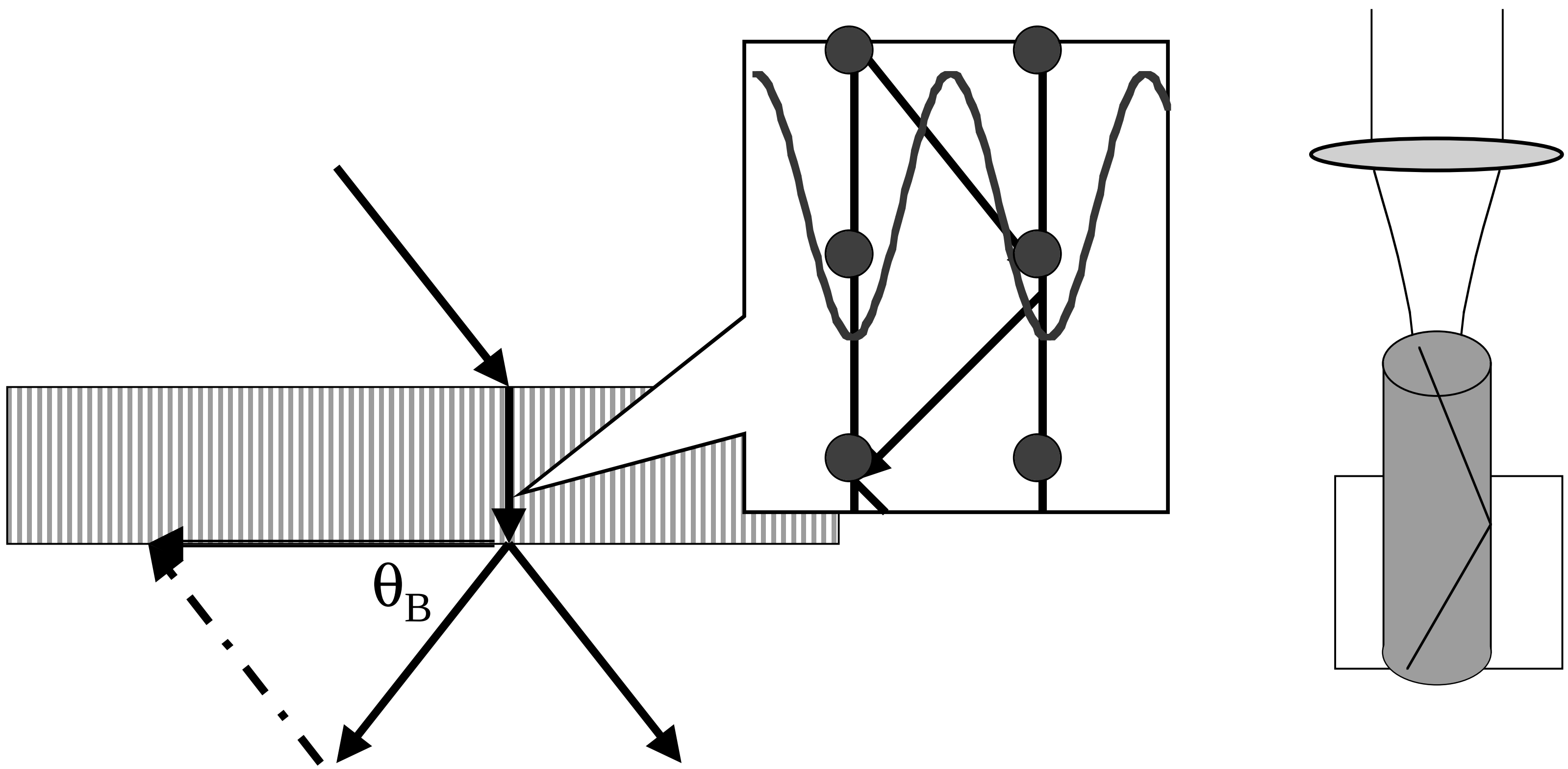
Simulation:
4 mJ/cm^2,
44 μrad/100 ps convolution



Borrmann Effect:

(2 r’s,2 n’s, 2 f’s 2 e’s)

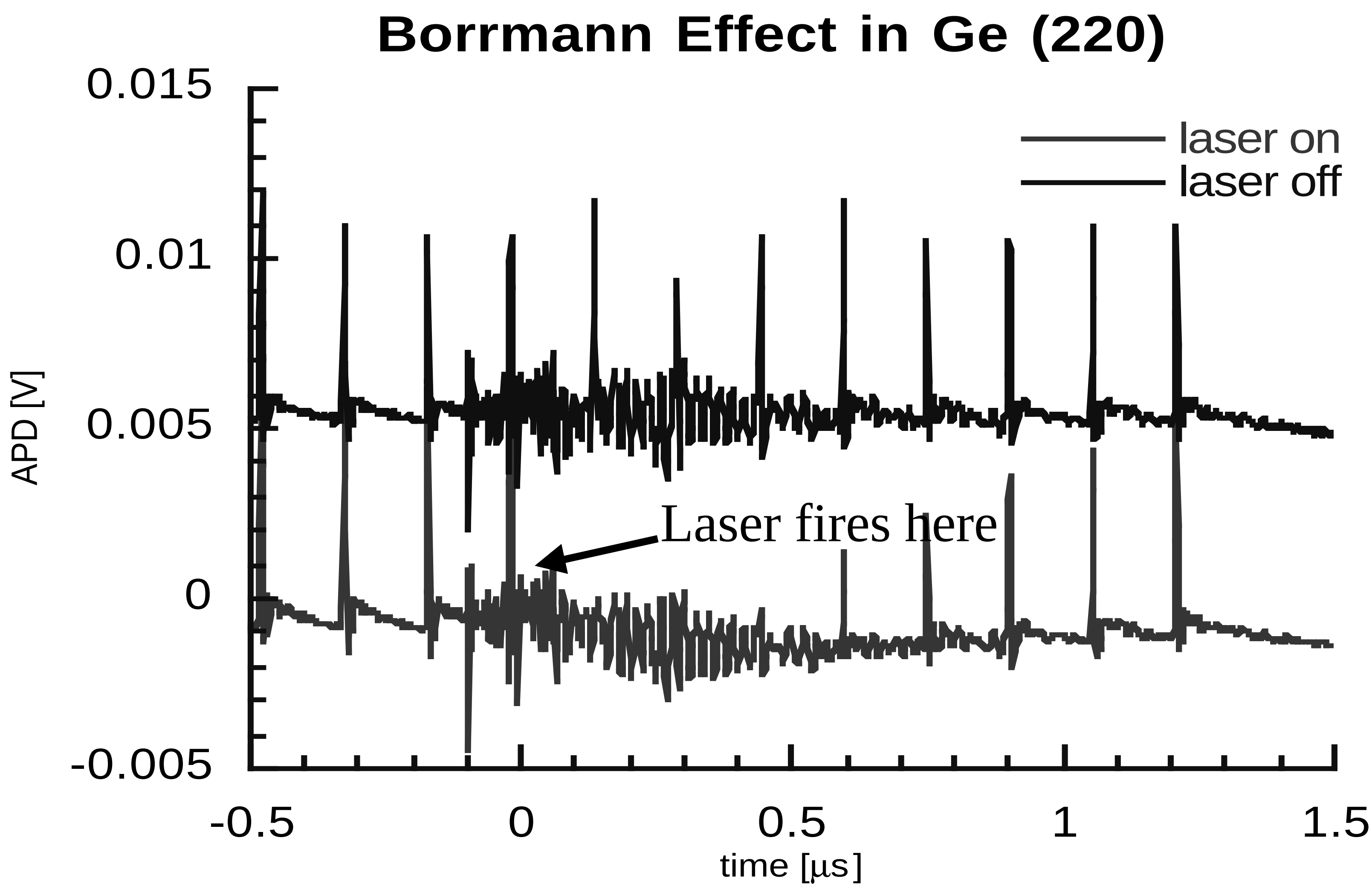
An X-ray waveguide



Anomalous Transmission

X-ray propagates with little loss over many incoherent-Absorption depths: Extended Probe!

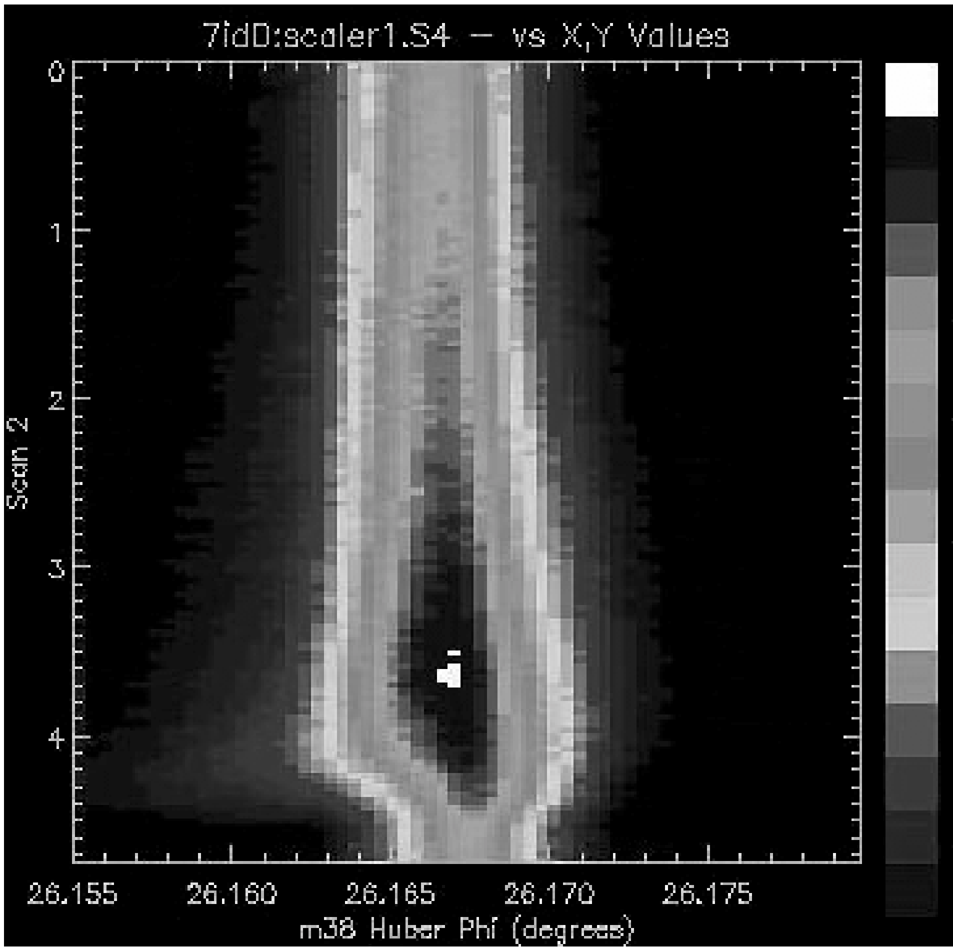
Bunch Pattern Shows long term effect of laser-heating on lattice

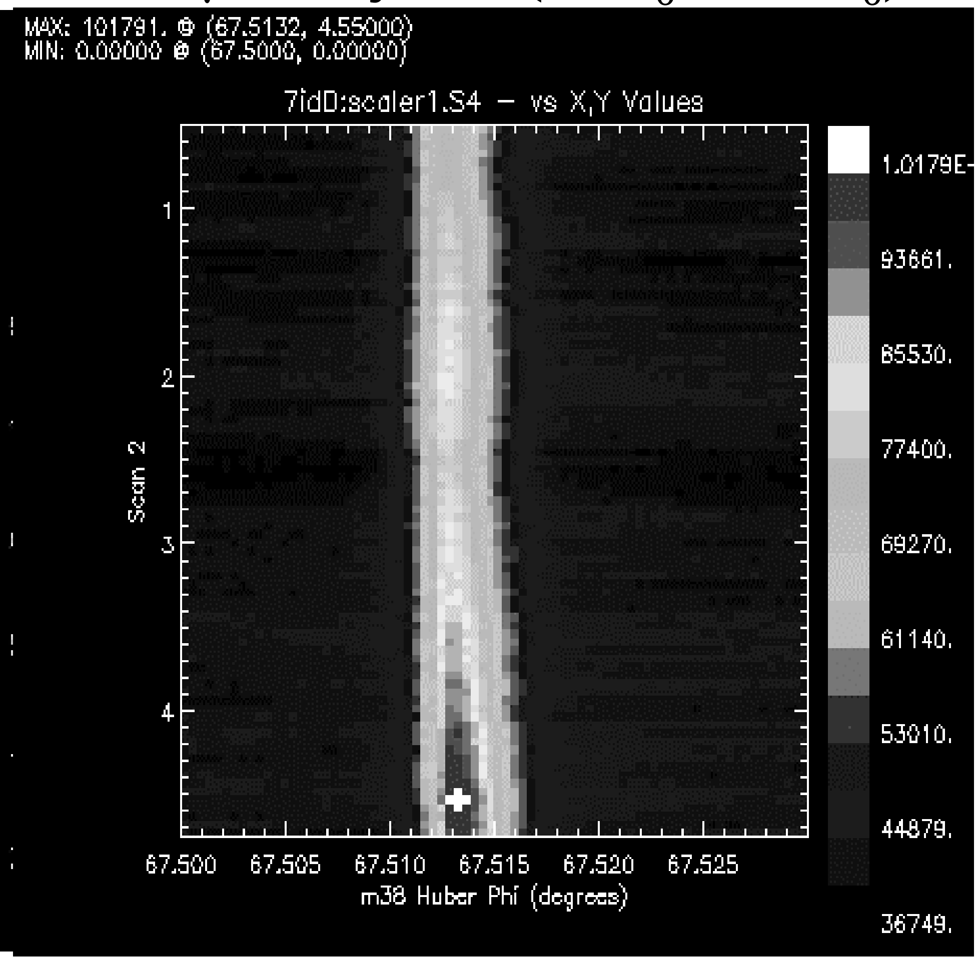


Acoustic Phonons in Ge

(004) Symmetric Bragg

(220) Symmetric Laue (Borrmann)
635 μm Crystal. ($13\ x_0$->$0.3\ x_0$)



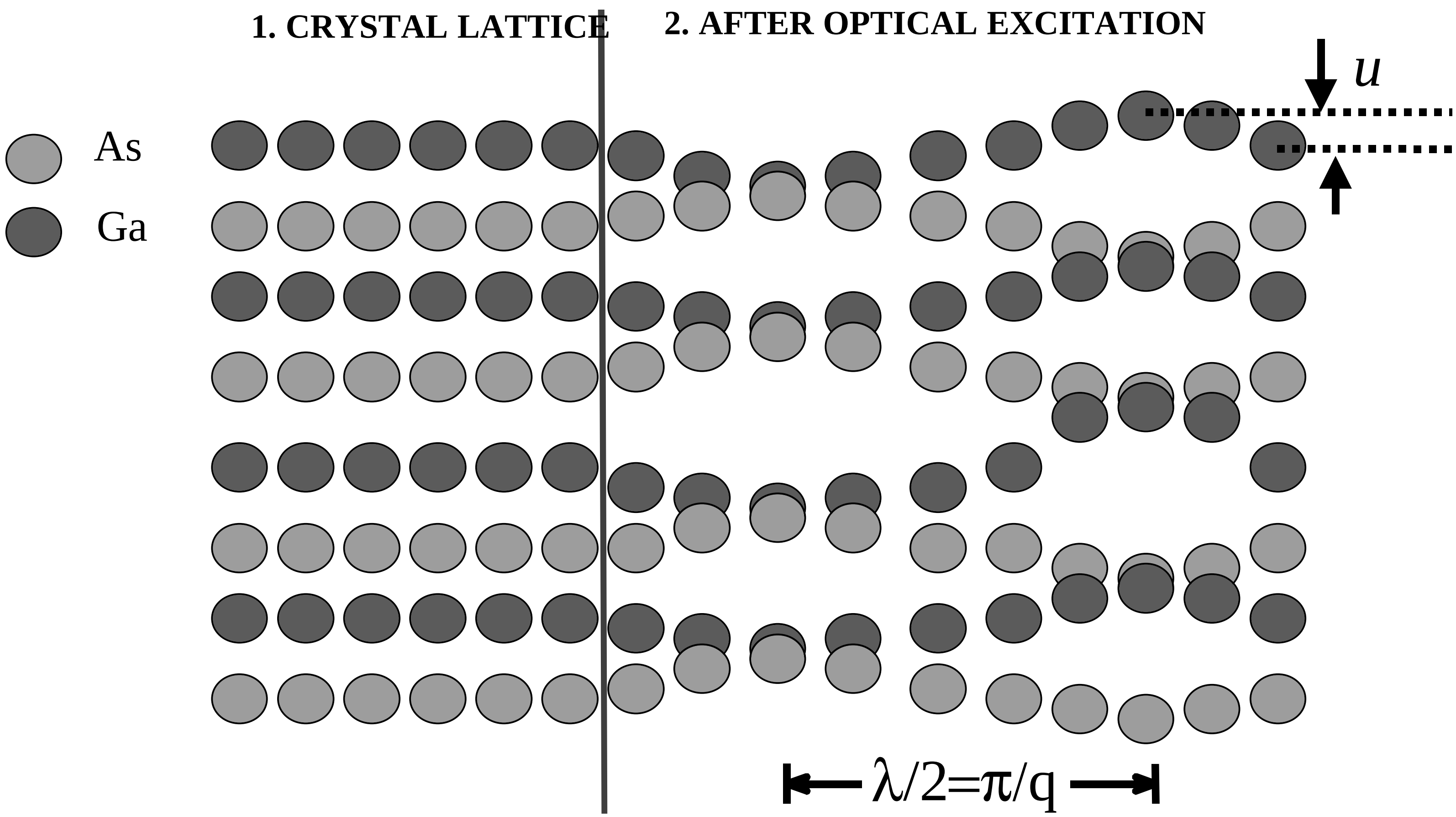


Doubly Anomalous Transmission?

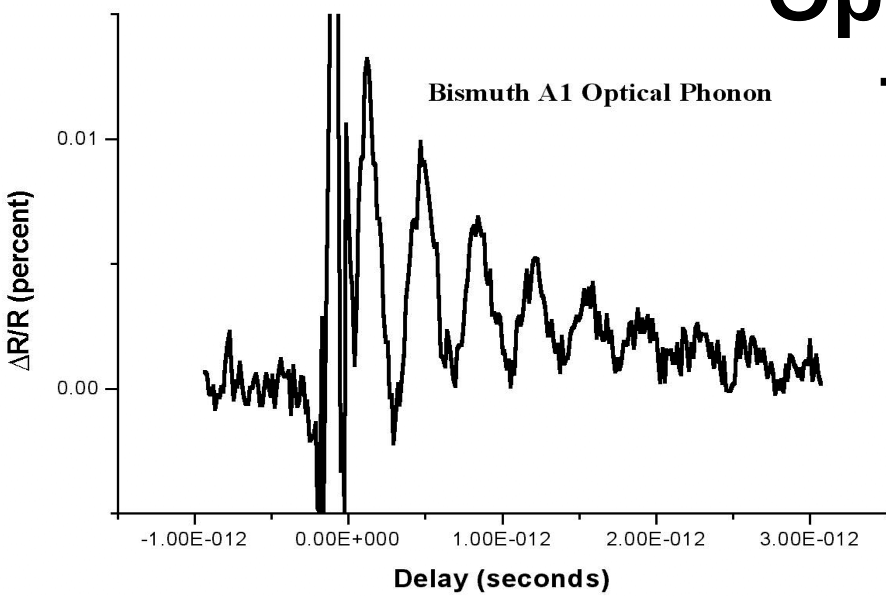
Coherent Optical Phonons

- Near single frequency: Can coherently switch on and off.
- Optical phonons typically in the 3–12 THz range: <50 fs switches possible.
- Create switch by inducing an optical phonon super-lattice with $k \sim 1/\lambda_{laser}$.
- Or, In “Forbidden” and “Weak”Reflections can use $k \sim 0$ phonons.

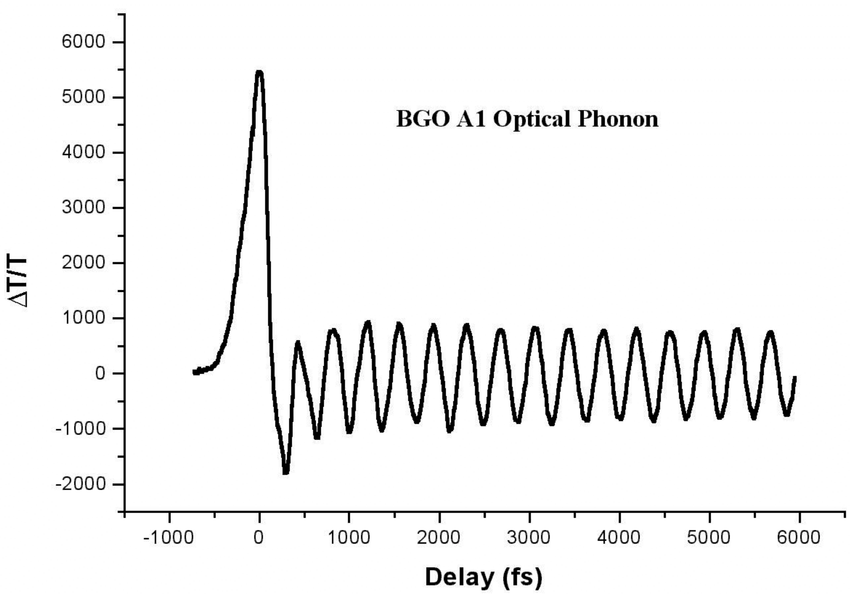
Optical Phonon Standing Waves



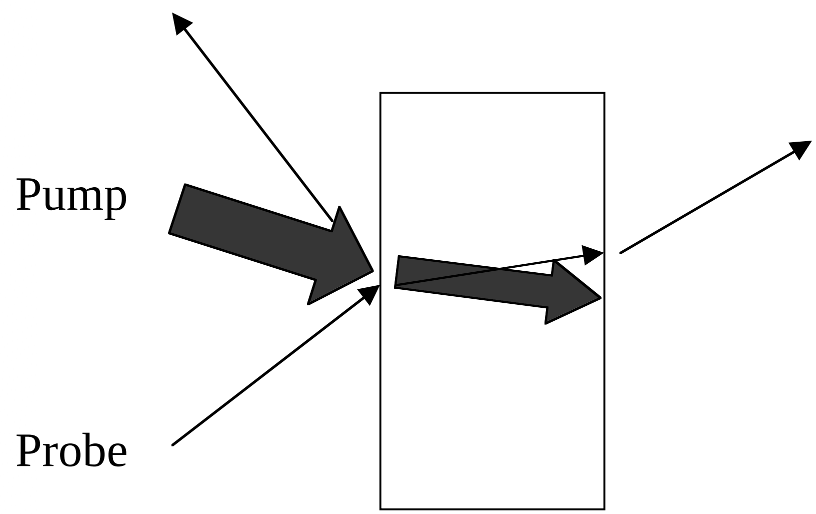
Optical Pump-Probe data for Bi and $Bi_4Ge_30_{12}$



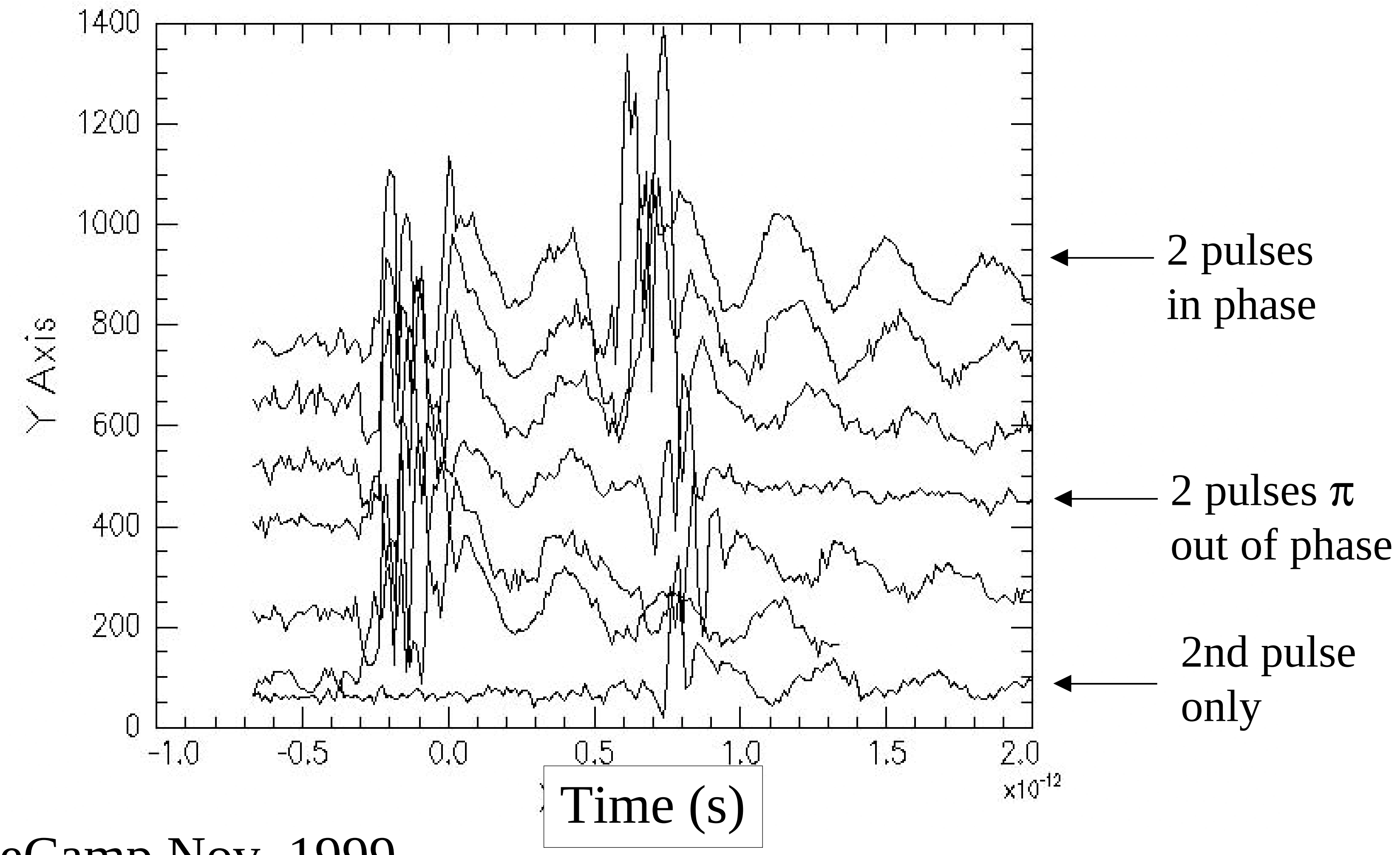
The bismuth data is showing the change in surface reflectivity while the BGO data is showing the change in bulk transmission.



Experimental setup



Coherent Control of Optical Phonons in Bismuth
Lattice distortions > 1%

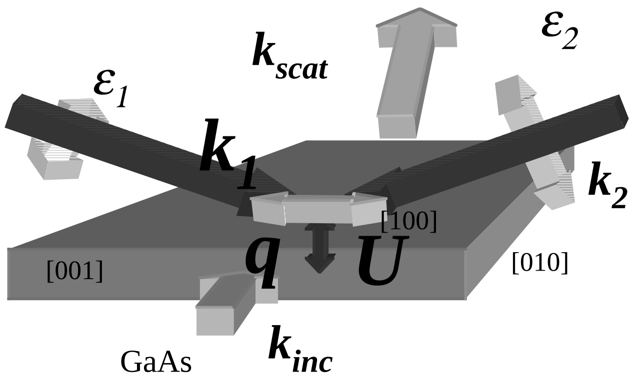


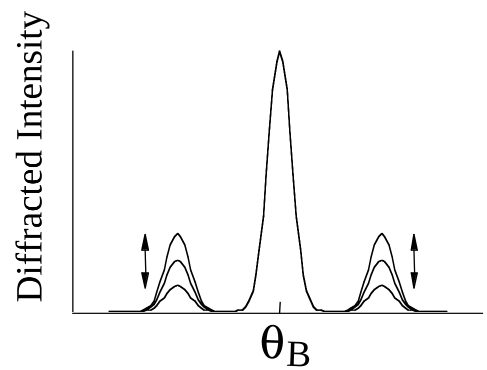
M. DeCamp Nov. 1999

Progress towards an Ultrafast Bragg Switch for X-Rays

Goal: To produce a sub-picosecond switch for X-rays.

Transient Grating: Create coherent superlattice using optical phonons
And Change the Bragg condition

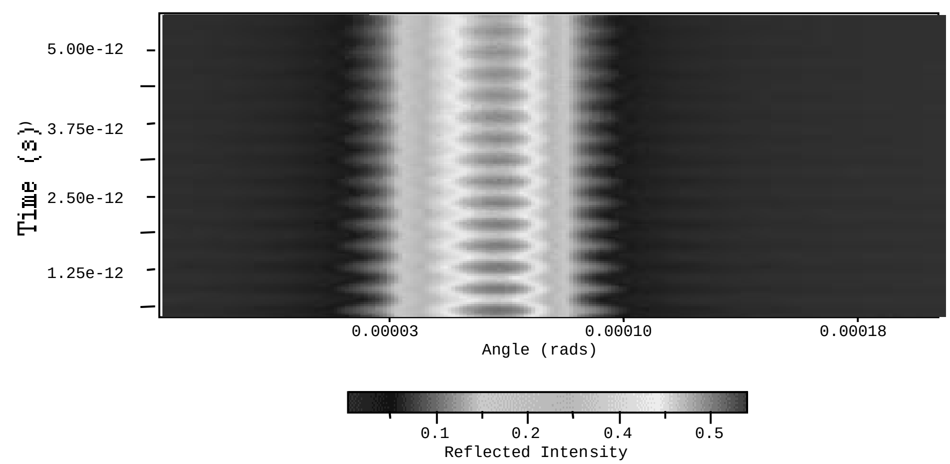




Modify structure factor using coherent optical phonons

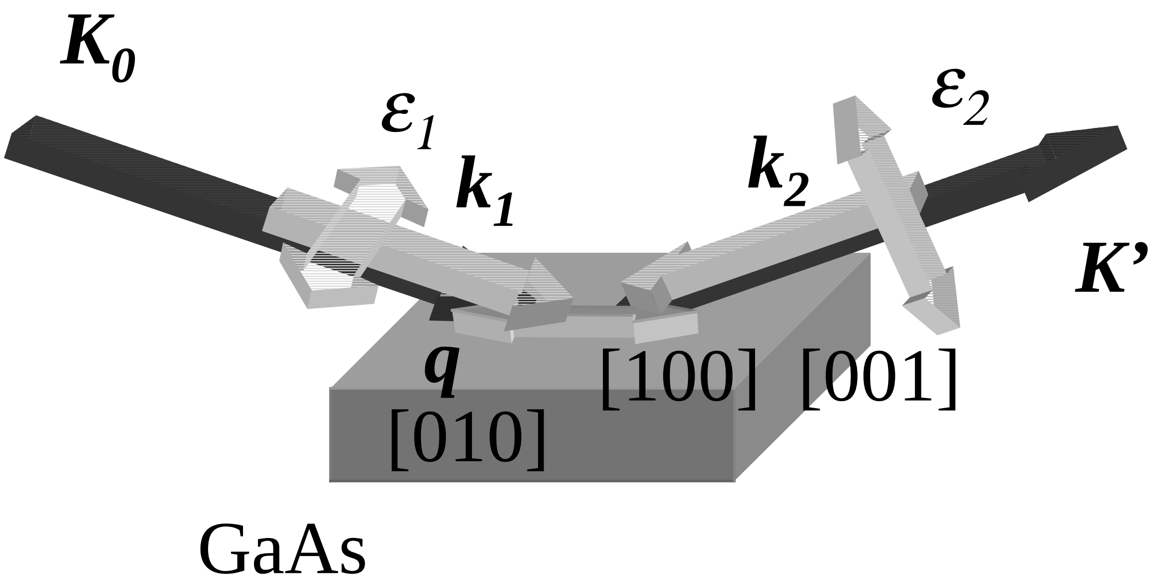
Simulation of (0003) Bismuth
Oscillations = 10^{-3} lattice shift

Oscillations of 10^{-2} have been measured using optical techniques



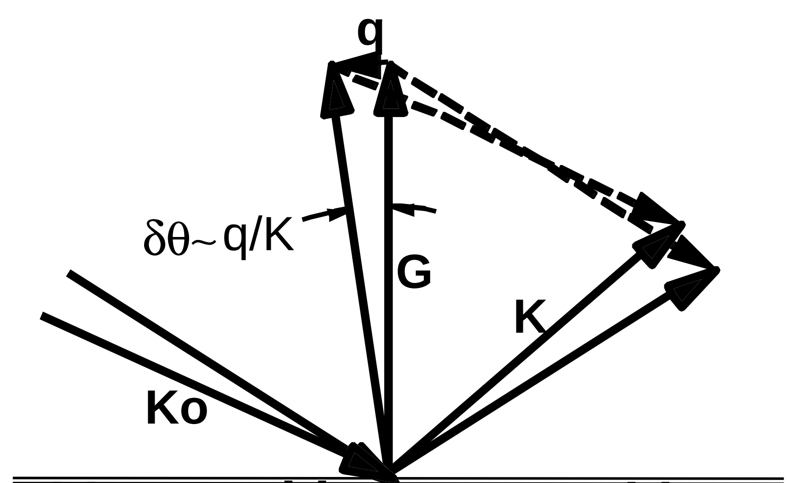
This project is supported by DOE-BES

The added periodicity of the lattice changes the Bragg Condition



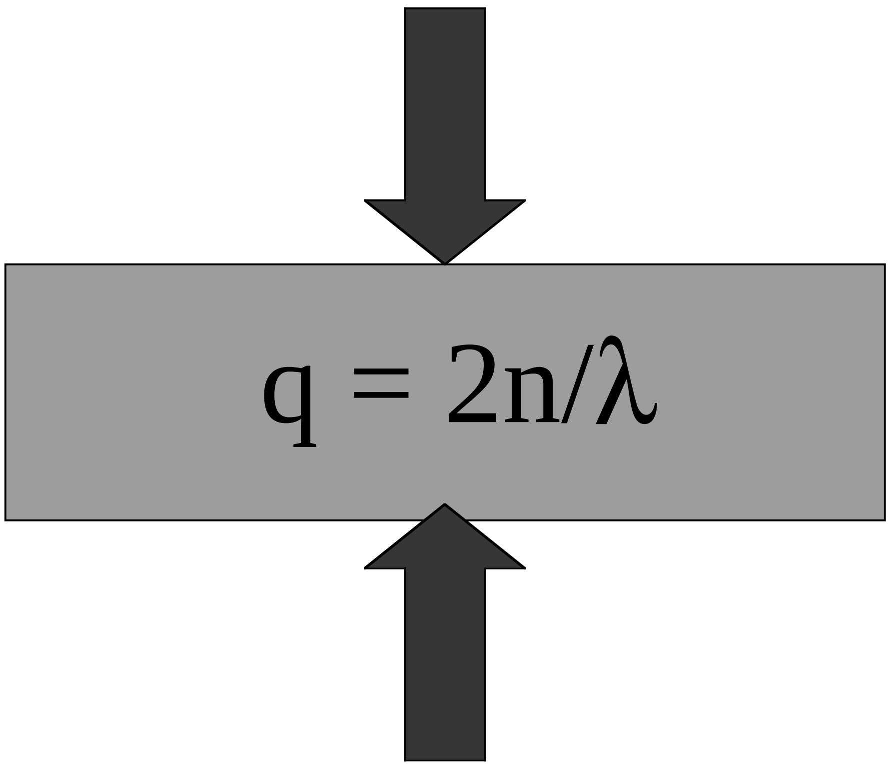
2 sets of Laser pulses:
One to set up standing wave;
One to destroy it.

Requires high quality single x-tals,
Large number of fringes (temp overlap)



$$\vec{K}_0 + \vec{G} + \vec{q} = \vec{K}'$$

In Transmission can generate higher wavevectors



Again temporal overlap and
Number of fringes
Becomes a critical issue

In k=0 optical phonons, a forbidden reflection can become allowed for modes where the crystal symmetry is changed

or for reflections that are "accidentally forbidden"

$$F_{hkl} = \sum_n f_n e^{2\pi i \vec{K} \bullet \vec{r}} = 0$$

$$r \to r + \partial r$$

$$F_{hkl} \approx \sum_n f_n 2\pi i \vec{K} \bullet \partial \vec{r} \neq 0$$

Where (hkl) are the usual Miller Indices

Could this work with strong reflections in the Laue geometry!?

Where are we headed from here?

- Detailed studies of strain and melting
- Optical phonon generation and ultrafast switches (BGO?)
- MQW (folded acoustic waves) studies
- Coherent effects at grazing incidence